



The role of precursors on the stabilisation of jet flames issuing into a hot environment



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ABSTRACT

This paper seeks to address unusual flame stabilisation behaviour observed in experimental jet flames which issue into a hot coflow. It has been observed that increasing the temperature and/or oxygen concentration in the coflow can lead to an increase in flame liftoff height. The paper isolates the role of chemistry, and in particular flame intermediates, on the observed phenomenon with a view to better understand how the behaviour changes over a range of conditions. A descriptive theory for this behaviour is proposed, which is based on the well-established theory that a build-up of radicals and intermediate species is responsible for autoignition of these flames. This paper systematically examines the role of these precursors with a view to better understanding of the chemical kinetics and to assess if the observed behaviour is chemistry-dominated. To this end, laminar flame calculations and ignition delay curves are presented, and the findings are validated with experiments. The results indicate that chemical effects alone are insufficient to fully explain the observations, but the calculations support the general trends noted in the experiments and highlight the importance and relative effects of some key precursors. In particular, the production and consumption of formaldehyde in a low oxygen environment supports the unusual flame behaviour observed experimentally.

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1. Introduction

Lifted flames issuing into hot environments have received much attention over recent years because of their relevance to practical systems that incorporate recirculation of hot combustion products [1–5]. Many studies have been performed to investigate various aspects of these flames, such as autoignition (e.g., [2,3]) and developing fundamental-level understanding of the MILD (moderate or intense low oxygen dilution) combustion regime without the complex flow recirculation typically encountered in a practical system (e.g., [4,5]). The current paper reviews flame stabilisation mechanisms specifically for flames issuing into a hot environment, with a focus on the effects of temperature and oxygen level. More general review articles on lifted flames have recently been compiled by Lawn [6] and Mastorakos [7].

To replicate the conditions of a fuel jet issuing into a hot coflow, the vitiated coflow burner (VCB) was developed by Cabra et al. [2,3] for the study of lifted flames. DeVised independently of the VCB, the jet-in-hot-coflow (JHC) burner was designed by Dally et al. [4] to investigate the detailed reaction zone structure of MILD combustion. Both of these burners feature a central fuel jet which issues into a coflow of combustion products from a secondary

premixed flame. The coflow temperature and composition are controlled by the operating conditions of the secondary burner. Whilst the flames in the JHC burner experiments of Dally et al. [4,5] were attached to the jet (as evident by measurements of OH and major species) the similarity of the operating conditions with the VCB makes it ideal to compare the differences in the flame behaviour across a range of conditions. It should be noted that although MILD combustion is sometimes referred to as “flameless” combustion, the behaviour of the reacting jet is consistent with the conventional definition of a turbulent non-premixed jet flame.

Using the VCB a sensitivity of liftoff height to small changes in the coflow temperature has been observed experimentally with hydrogen/nitrogen and methane/air fuels in the jet. Analysis has revealed a build-up of radicals and intermediate species prior to ignition of these flames and indicates that autoignition is the stabilisation mechanism of these flames [8–11]. The evolution of these autoignition kernels is known to be a function of the balance between radical production and dissipation [12]. Extensive modelling activity has been conducted to replicate this phenomenon. Much of this modelling, however, has concentrated on the ability to replicate the observed trends, rather than understanding the governing physics. Notwithstanding the highly sensitive influence of small variations in coflow temperature on the liftoff height, systematic studies in the VCB have typically concentrated on a narrow range of conditions, corresponding to the experimental

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measurements. Whilst some insightful systematic studies have been performed (e.g., [12]) these have been limited to conditions very close to the two experimental studies of Cabra et al. [2,3]. In contrast, the JHC burner has been used over a much wider range of conditions, including both MILD combustion and autoignited flames [13]. However, there still exists significant gaps in understanding the flame behaviour across a range of operating conditions, which is one of the objectives of the current paper.

In experimental studies using the JHC burner by Medwell et al. [14] lifted flames were achieved in the transition to MILD conditions. These flames were seen to be very different from conventional lifted flames, including the presence of both CH_2O and OH upstream of the liftoff height. This observation led to them being referred to as “transitional” flames, where a transition in appearance is noted at the apparent liftoff height. Further investigation of these flames also revealed counter-intuitive results. Reducing the O_2 level in the coflow initially led to an increase in liftoff height (as may be expected), but further reducing the O_2 level causes the flame to re-stabilise to the jet exit plane [13]. More recently, the trend of increased liftoff with increased coflow O_2 level has been shown in numerical experiments [12,15]. Worth noting is that changes in the O_2 level of the coflow can affect the flame entrainment ratio, with a reduction in O_2 level increasing the entrainment ratio [16]. Decreasing the coflow temperature level also caused a similar trend, where the liftoff height was seen to initially increase (as may be expected) before the liftoff height starting to decrease with lower coflow temperature [13]. Although observed experimentally, a detailed description of these phenomena is not fully understood. Reaction rate analysis of hydrogen/nitrogen flames, however, indicates that the hydrogen radical (H) may play an important role in explaining the observed behaviour [12]. In the case of hydrocarbon flames in general, other radicals and intermediate species have an influential role in the stabilisation mechanism.

Imaging measurements of lifted flames in a hot coflow show a build-up of formaldehyde (CH_2O) upstream of the flame liftoff height, after which the concentration of OH rapidly increases with downstream distance [10,14]. In both the VCB and the JHC burners it is widely accepted that the presence of CH_2O and other intermediates upstream of the liftoff height is evidence of autoignition [7]. Due to the different coflow temperature and O_2 levels the appearance of the OH and CH_2O upstream of the liftoff height is quite different between the measurements of Gordon et al. [10] and Medwell et al. [14]. In the autoignitive flames in the VCB, Gordon et al. [10] show small pockets of isolated OH indicative of spontaneous ignition kernels. In contrast, in the JHC burner the flames approaching MILD combustion show continuous OH measurements upstream of a transition to the fully burning OH concentration [14]. Nonetheless, in both cases the high temperature coflow is responsible for the production of CH_2O in the lifted region of these flames. It is also noted that, despite the high temperature of the coflow ($>1100\text{ K}$), the temperature of these flames along the centreline is expected to be below $\sim 900\text{ K}$ which represents the initiation of $\text{CH}_2\text{O} \rightarrow \text{CO}$ oxidation [17], and thus any CH_2O that is formed is unlikely to oxidise in this region, leading to its accumulation. DNS results of a lifted ethylene flame support the experimental observations of precursor species (such as HO_2) upstream of the OH reaction zone [18].

The autoignition process is well-known to initiate and stabilise through the build-up of radicals [19,20]. For hydrogen flames in a hot coflow HO_2 is considered an important precursory species [21,22], and is believed to trigger autoignition [23]. The presence of intermediate species are also known to reduce the ignition delay under MILD combustion conditions [24]. In particular CH_2O is known to be very important in the hot oxidant environment [25], and is expected to aid ignition [26]. The affect of OH radicals in

the oxidant stream from a two-stage combustion processes is also expected to reduce the ignition delay [27].

Another important observation noted in the JHC burner is a reduction in apparent liftoff height with an increase in jet Reynolds number [14,28,29]. This is not seen in the autoignitive lifted flames of Cabra et al. [2,3]. It is reported that an increase in turbulence delays autoignition, but once a flame is established the increased mixing enhances the overall rate of reaction [30]. Increasing the turbulence intensity in the JHC flames reduces the apparent liftoff height, rather than increasing it, indicating that the combustion reaction had already initiated. These observations support the previous assertion that these flames are not autoignitive lifted flames, but rather MILD or transitional flames [14]. A further factor that may contribute to the observed flame behaviour is the differences in mixing due to laminarisation that can occur due to the different temperatures, both from the coflow and the reaction zone heat release [31].

In summary, flames in a hot environment may exhibit different behaviour, dependent on the properties of the coflow. It is widely accepted that autoignition is responsible for stabilisation of lifted flames in a hot coflow [12], however, with reduced O_2 levels in the coflow the flame behaviour is observed to be quite different [13]. It is postulated that the high temperature oxidant stream promotes a radical-pool build-up which assists in the stabilisation of these flames. The role of O_2 can have pronounced differences, however, on the flame stability. A descriptive process of stabilisation theory for lifted flames in a hot coflow is proposed as follows:

- With a (relatively) high coflow temperature and O_2 level lifted flames under non-MILD conditions stabilise through autoignition. (The precise definition of *relatively* high depends on the operating conditions, but loosely may be considered $\geq 10\% \text{ O}_2$ and $\geq 1400\text{ K}$.) The high temperature of the coflow initiates precursory reactions and builds a pool of radicals and minor species. The pool of precursors is expected to induce and promote rapid ignition. This is consistent with the summary provided by Gordon et al. [8–10].
- Reducing the coflow temperature and/or O_2 level of a lifted flame leads to an increase in liftoff height, as may be expected, due to a reduction in reaction rates.
- Further reducing the coflow temperature and/or O_2 level leads to flames in the MILD combustion regime. These flames typically attach to the jet exit plane, despite the lower temperature and O_2 level. This is due to O_2 penetration across the reaction zone to the fuel-rich side, which further enhances the precursor pool under these conditions, and is particularly prevalent under high strain conditions [32]. It is apparent that MILD conditions promote the production of a precursor pool leading to more stable flames that are less prone to liftoff. Increasing the jet velocity further stabilises the flame, both through increased O_2 penetration and enhanced mixing.

The aim of the present paper is to review and analyse the proposed theory regarding flame stabilisation of lifted flames in a hot coflow and assess flame behaviour across a range of conditions, highlighting the importance of minor species. Notwithstanding the important role of turbulence-chemistry interactions (which can enhance or inhibit autoignition [33–35]) and jet autoignition in heated air [36], these scenarios are beyond the scope of the present work which focuses on chemical-kinetic effects in a hot coflow at various O_2 levels. It is believed that stabilisation mechanisms in the hot coflow environments are predominately affected by the chemistry [23], supported by the observation in practical combustors employing these types of flames are only weakly dependent on operating conditions [25].

2. Methodology

The Chemkin software package is used throughout this paper to assess the influence of chemical kinetics on the reaction zone structure and ignition delay time of the flames of interest. Ignition delay has been suggested as a good measure of the characteristic time-scale for autoignition and MILD combustion [35,37,38].

The fuels considered in this paper are either methane (CH_4), ethylene (C_2H_4), or a mixture of natural gas (NG) and hydrogen (H_2) in an equal volumetric ratio. For the NG/ H_2 case, natural gas is assumed CH_4 (noting that the corresponding experiments used natural gas with $\geq 92\%$ CH_4). In all cases the GRI-Mech 3.0 chemical kinetic mechanism is used.

For each fuel type, calculations are performed for a range of oxidant stream compositions, and over a range of oxidant stream temperatures, including experimental conditions reported in the literature. The key oxidant compositions are summarised in Table 1. The O_2 level for each composition is different, and hence this serves as a unique identifier. The 3% and 9% O_2 cases are representative of the coflow composition from the experiments conducted by Medwell et al. [5,14]. The 12% O_2 case corresponds to the coflow used by Cabra et al. [3]. Also included is standard air, assumed N_2 and O_2 only. For the different experimental conditions reported in Table 1 the CO_2 and H_2O concentration in the oxidant stream is not constant. In particular, it should be noted that the Cabra et al. coflow is from a lean H_2 /air flame, and so will have a different composition to the CH_4 / H_2 / N_2 / O_2 flame used by Medwell et al. for the 3% and 9% O_2 cases. Other minor species may also be different between the various coflow conditions in experiments, however, minor species in the oxidant stream are not included in the calculations. For a fixed O_2 level, the differences in the composition of the coflow have previously been shown to affect the liftoff height [13]. Nonetheless, throughout this paper most comparisons are made between the 3% and 9% O_2 cases (for which the CO_2 and H_2O concentration is constant). Comparisons between the other cases are to highlight the trends observed experimentally, and thus the influence of the inert concentration is not critical.

To investigate the influence of strain on opposed-flow laminar flames, the OPPDIF routine of Chemkin is used with a view to assessing the effect on the precursory species. Also considered is the effect on the reaction zone structure by specifically adding precursory species to the fuel stream. The strain rate on the reaction zone is varied by changing the inlet velocity of the fuel and oxidant streams. The strain rate reported in this paper is the peak normal strain rate. The temperature of the oxidant stream is also varied, but the fuel stream temperature is fixed at 300 K for all calculations, similar to the experiments. Multi-component transport is used to account for preferential diffusion effects which are known to be important under these conditions [39,40].

To determine the ignition delay time for various stoichiometric mixtures of fuel and oxidant (as per Table 1) the AURORA routine of Chemkin is used. The ignition delay is taken from the AURORA post-processor and is determined on the basis of maximum rate of change of temperature. Autoignition is known to occur at the location of the most reactive mixture [35,41–44]. It should be

noted that the most reactive mixture (having minimum ignition delay time) is likely to occur under lean conditions for the case of a cold fuel with a hot oxidant stream [19,20]. Calculations were also performed over a range of equivalence ratios, however the general trends are the same as for stoichiometric conditions, and not included in this paper. The ignition delay is used as an indicator of the level of reactivity of the various fuel/oxidant mixtures at different initial temperatures without the effect of mixing or diffusion.

3. Results

3.1. Laminar opposed-flow flame calculations

Formaldehyde and precursors are formed upstream of the liftoff height in lifted flames in a hot coflow and it is postulated that these precursory species contribute to the stabilisation of these flames. In attached jet flames, Medwell et al. [32] have shown that formaldehyde is also a significant intermediate under MILD combustion conditions, where it is produced on the fuel-rich side as a consequence of O_2 penetration across the reaction zone [13]. To investigate the role of formaldehyde of jet flames in a hot coflow in a general sense, laminar flame calculations are presented in this section to demonstrate some features of flames in a hot coflow, and how these differ from conventional flames.

The OPPDIF routine of the Chemkin package is used to study the detailed structure of opposed-flow diffusion flames. This configuration is analogous to the one-dimensional traverse across the reaction zone of a non-premixed flame. Results are presented for a range of imposed strain rate, which is varied by changing the velocity of the fuel and oxidant streams. The strain rate shown is the maximum strain rate encountered across the reaction zone.

Figure 1a shows the peak temperature from the laminar flame calculations for the conditions experimentally investigated by Medwell et al. [5] of attached flames in a JHC burner. The fuel is a mixture of natural gas (assumed methane) and hydrogen in equal volumetric ratio issuing into a coflow of hot combustion products at 1100 K with either 3% or 9% O_2 concentration. The coflow composition is presented in Table 1. For comparative purposes, also shown in Fig. 1a are the results for air at 21% O_2 at an inlet temperature of either 300 K or 1100 K. The peak temperature with 3% O_2 is lower than the other conditions across the range of strain rates considered. This observation is consistent with the expected features of MILD combustion. Of particular note is that the flames with a hot oxidant stream (1100 K), but low O_2 level (3% or 9%), are able to withstand a much higher strain rate before extinction, extending well above 2000 s^{-1} . The dominant role of initial temperature over the oxidant O_2 level is well-known [45]. Importantly, however, for the case with 21% O_2 air, even at a temperature of 1100 K, the laminar flame calculations predict extinction at a lower strain rate than for either the 3% or 9% O_2 cases ($a \sim 1500 \text{ s}^{-1}$). From a simplistic perspective this appears counter-intuitive that higher O_2 levels lead to a lower critical strain rate, however, this observation is consistent with the proposed theory of stabilisation under low O_2 conditions [32].

Figure 1a also includes the results for the methane/air flame conditions of Cabra et al. [3] in the VCB. It is apparent that despite the higher oxidant stream temperature and O_2 level of this flame, the laminar flame calculations predict that it extinguishes at a lower strain rate than the flames under MILD conditions. This is partially explained by the fact that these flames contain air in the fuel stream rather than H_2 , which is expected to improve the flame stability. Importantly, however, these results support the experimental observations that the 12% O_2 flames appear lifted, whilst the flames are attached with a lower O_2 and lower temperature

Table 1

Oxidant stream composition from Medwell et al. [5] for 3% and 9% O_2 coflow. 12% O_2 case from Cabra et al. [3]. Also included is air (21%). Normalisation is performed on a volumetric basis.

O_2 (%)	N_2 (%)	CO_2 (%)	H_2O (%)
3	84	3	10
9	78	3	10
12	73	0	15
21	79	0	0

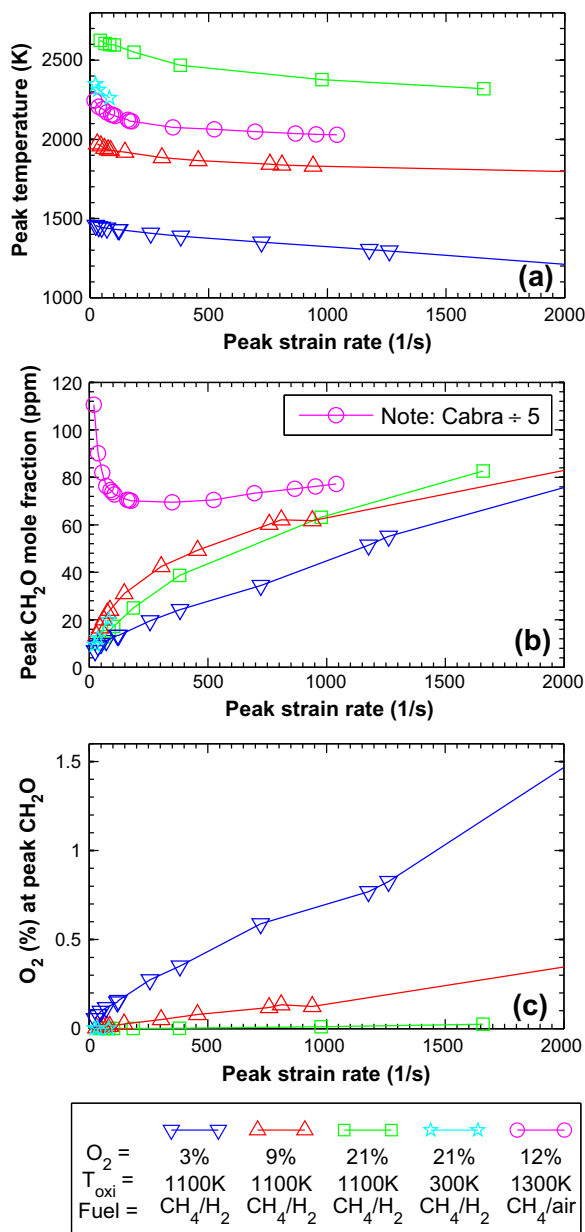


Fig. 1. Strained laminar flame calculation results: (a) peak temperature; (b) peak formaldehyde (CH₂O) mole fraction (note that Cabra flame results are scaled by a factor of five); (c) O₂ mole fraction at the location of peak CH₂O. Fuel and oxidant temperature & O₂ as shown in legend, with the balance as per Table 1.

oxidant. It should be noted that the factors governing liftoff in a turbulent jet flame extend beyond those modelled in simplified laminar flame calculations. Nonetheless, the strained flame calculations give an indication of the propensity of a flame to liftoff due to strain.

Figure 1b shows the peak formaldehyde (CH₂O) mole fraction as a function of strain rate for the same conditions as Fig. 1a. Unlike the temperature (and OH, not shown), the CH₂O is seen to increase with strain for the CH₄/H₂ flames. The peak formaldehyde concentration is also of a similar magnitude for all of the oxidant stream conditions. The CH₂O mole fraction results shown in Fig. 1b for the methane/air VCB flame conditions has been re-scaled by a factor of five to account for the much higher CH₂O concentration in this flame due to the partial premixing of the fuel stream with air. Although the fuel composition is very different for the VCB flame it is included to demonstrate that it has a very different profile

with respect to the imposed strain rate. An initial steep decrease in CH₂O concentration with increased strain, after which it almost plateaus is in contrast to the CH₄/H₂ flames. It may be expected that this is a contributing factor to the high sensitivity of the liftoff height to the coflow temperature of this particular flame, although this investigation is beyond the scope of the current work.

Following the approach adopted previously [32], Fig. 1c presents the O₂ concentration at the location of peak CH₂O mole fraction. Recall that CH₂O only occurs on the fuel-rich side of the reaction zone. These results clearly demonstrate the effect of reaction zone weakening under MILD conditions: despite a low O₂ level in the oxidant stream, a high O₂ concentration is found to occur at the location of peak CH₂O. To highlight this point, note that the fuel stream does not contain any O₂, yet for the MILD case (3% O₂) at the location of peak formaldehyde (i.e., on the fuel-rich side) the O₂ concentration is over half the O₂ concentration in the oxidant stream itself, at strain rate $\sim 2000 \text{ s}^{-1}$. At lower strain rates the O₂ concentration is relatively low, illustrating that these effects are sensitive to the strain rate of the flame. Note that Fig. 1c does not include the results of the Cabra et al. [3] CH₄/air flame because the O₂ distribution is not comparable with the other flame conditions on account of partial premixing.

At low oxidant O₂ levels, as the strain is increased the amount of O₂ that penetrates the reaction zone increases (as evident from Fig. 1c). This seems to be a mode of flame stabilisation under these conditions. The presence of CH₂O on the fuel-rich side seems to aid the chemical reactions and allows these flames to withstand much higher strain rates. Importantly, however, for the 21% O₂ case the level of O₂ penetration is significantly lower. This is consistent with the trend of extinction occurring at a lower strain rate and provides further evidence that reaction zone weakening effects being most dominant under MILD combustion conditions. These observations highlight the importance of CH₂O, especially under hot and diluted O₂ conditions.

To assess the relative effects on a different fuel, Fig. 2 presents the peak CH₂O mole fraction as a function of strain rate for ethylene (C₂H₄) fuel over a range of oxidant temperature and O₂ levels. Consistent with the reaction zone weakening effects noted from the CH₄-based fuels, as either the oxidant O₂ level or temperature is decreased, the CH₂O concentration increases for C₂H₄ as well. The CH₂O is also noted to increase with imposed strain rate, again as a result of reaction zone weakening. Coupled with the

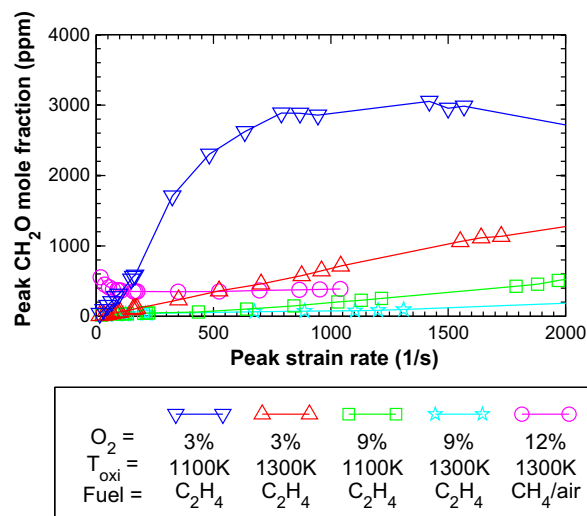


Fig. 2. Strained laminar flame calculation results of peak formaldehyde (CH₂O) mole fraction. Fuel and oxidant temperature & O₂ as shown in legend, with the balance as per Table 1.

observation that the maximum strain rate achievable prior to extinction is accompanied by an increase in the peak CH_2O level, the results presented in this section support the proposed flame stabilisation theory.

3.2. Ignition delay

The opposed-flow laminar flame calculations are limited to the analysis of attached flames. Whilst the proposed theory seems to explain the stabilisation of the attached flames in a hot coflow of Medwell et al. [5] and contrast these findings to those from the VCB [3], for analysis of lifted flames a more general approach is required. This section presents the results of ignition delay calculations, which are used to quantify the rate of reactivity of mixtures over the range of interest with a view to give an indication of the trends of lifted jet flames observed experimentally, whilst isolating the effects of mixing.

3.2.1. Oxidant O_2 and temperature

For the CH_4/H_2 fuel considered in Section 3.1 the ignition delay results for a stoichiometric mixture over a range of different oxidant stream temperature and O_2 levels is presented in Fig. 3. The oxidant stream is comprised of a varying O_2 concentration, with H_2O and CO_2 fixed at 10% and 3%, respectively, and N_2 forms the balance.

As may be reasonably expected, each of the ignition delay curves presented in Fig. 3 decreases exponentially with inlet temperature (as confirmed from a plot on a logarithmic scale, not included for brevity). The absolute value for each O_2 concentration follows a monotonic trend, i.e., the lowest O_2 level has the longest ignition delay and gets progressively faster as the O_2 concentration is increased. Both of these results are anticipated: increased temperature and O_2 level are both expected to reduce ignition delay. However, for jet flames that occur in this range of oxidant conditions, a non-monotonic relationship between liftoff height and temperature/ O_2 level has been reported [13]. The results presented in Fig. 3 indicate that ignition delay itself is not responsible for the non-monotonic behaviour.

To further demonstrate the similarity of the ignition delay curves, Fig. 4 presents the same results, but normalised with respect to the maximum ignition delay for each O_2 level. The results for other fuels (CH_4 and C_2H_4) also show the same trends. The curves are all seen to collapse onto one another, providing further evidence that the non-monotonic relationship observed experimentally is not a simple function of the chemistry.

3.2.2. Formaldehyde addition

Given the demonstrated link between an increase in formaldehyde concentration and increased stability, Fig. 5 shows the

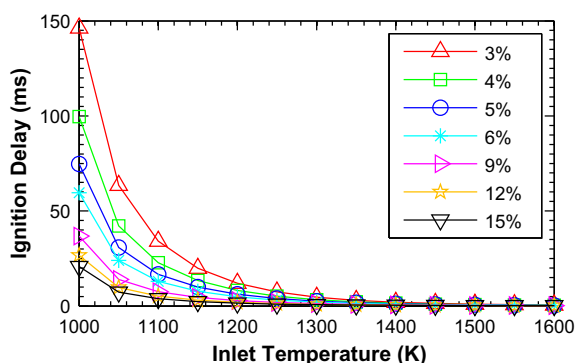


Fig. 3. Ignition delay (milliseconds) for stoichiometric CH_4/H_2 fuel across a range of initial temperatures and for different O_2 concentrations (3–15% O_2).

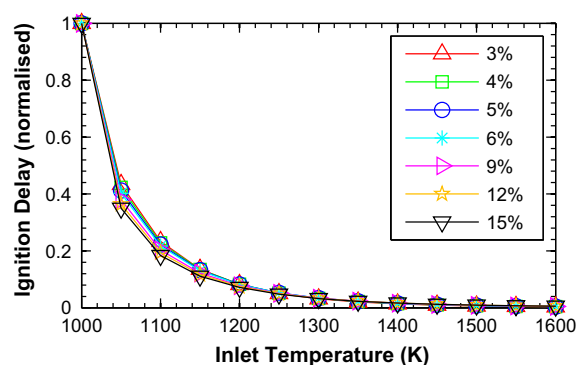


Fig. 4. Ignition delay (normalised to maximum ignition delay for each O_2 level) for stoichiometric CH_4/H_2 fuel across a range of initial temperatures and for different O_2 concentrations (3–15% O_2).

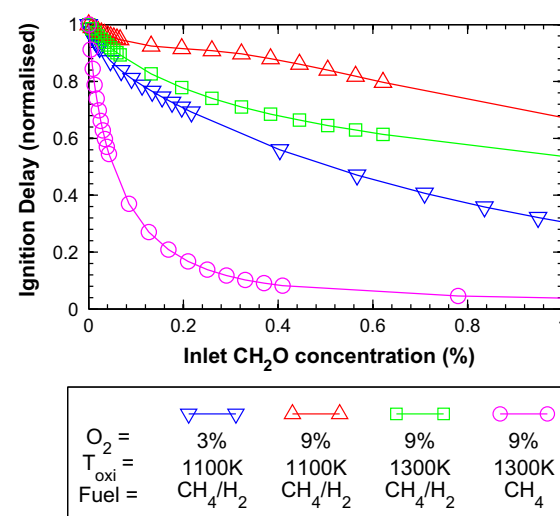


Fig. 5. Ignition delay (normalised to maximum for each condition) for various stoichiometric fuel/oxidant mixtures with the addition of CH_2O to the reactants.

influence of formaldehyde addition to the initial reactant mixture on the normalised ignition delay for stoichiometric mixtures for various fuels, O_2 levels in the oxidant and oxidant temperatures. Results at other stoichiometries ($\Phi = 0.8$ and 1.2) show similar trends and are omitted for brevity. The results for each fuel/oxidant mixture are normalised with respect to the case with no formaldehyde addition. This normalisation enables the relative trends of formaldehyde to be isolated for each of the flame conditions. The intention is to provide a replication of the trends observed experimentally, rather than relating absolute ignition delay from laminar calculations to the turbulent lifted flames observed experimentally.

In the attached flame MILD combustion case from Section 3.1 (CH_4/H_2 fuel, with 3% O_2 at 1100 K) it is seen from Fig. 5 that the addition of CH_2O to the mixture reduces the ignition delay. For the same fuel and temperature, as the O_2 concentration is increased to 9% O_2 , the addition of CH_2O also reduces the ignition delay, but the affect seems less influential. This finding is consistent with the experimental observations that when the coflow O_2 level is increased from 3% to 9% that attached jet flames are seen to lift off. From Fig. 5, at 9% O_2 the mixture appears less responsive to the build-up of precursors (namely CH_2O) that occurs under the hot and diluted conditions. Whilst it should be noted that an increase in oxidant stream O_2 concentration has been observed to correlate with increased liftoff height [13] the CH_4/H_2 fuel considered in

Fig. 5 does not produce lifted flames. The general trend, however, of increased “reactivity” as a result of the presence of CH_2O is more important at lower O_2 conditions and consistent with Fig. 5. Note-worthy is that further increases to the inlet temperature (to 1300 K) recovers some of the reduced ignition delay with CH_2O addition at 9% O_2 concentration.

The general trends noted in the behaviour of the ignition delay with changes in the oxidant composition for the CH_4/H_2 mixtures support the general observations noted for other fuels. However, to highlight the role of fuel type on the observed behaviour, Fig. 5 also includes the results for CH_4 fuel. Without the additional H_2 the role of CH_2O on CH_4 flames is even more influential, possibly as a result of reduction of consumption of CH_2O via H_2 combustion routes, but the general trends remain consistent.

To assess the role of CH_2O addition on CH_4 flames over a wider range of oxidant temperature and O_2 levels, Fig. 6 shows ignition delay calculation results for a constant 0.5% CH_2O concentration in the mixture. The ignition delay presented is normalised to the case of no CH_2O addition with the corresponding composition and temperature. Towards the lower temperature limit the addition of 0.5% CH_2O significantly decreases the ignition delay – by more than a factor of 30, relative to the no CH_2O case. At higher inlet temperatures and O_2 concentration the influence of CH_2O on the ignition delay is less pronounced, but still significant.

In comparison to the case of CH_4 only, the CH_4/H_2 mixture results in Fig. 7 indicate that the addition of 0.5% CH_2O is less significant (consistent with Fig. 5). Nonetheless, for the low O_2 conditions, the addition of CH_2O is still observed to more than halve the ignition delay. This reduction is most pronounced with the lower O_2 levels, but a non-monotonic trend is noted with an increase in inlet temperature.

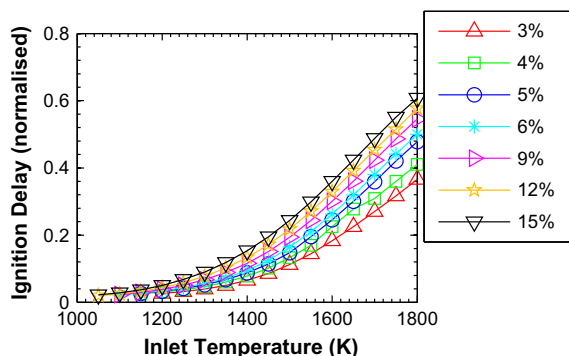


Fig. 6. Ignition delay (normalised to the no CH_2O case) for stoichiometric CH_4 with 0.5% CH_2O added across a range of inlet temperature and O_2 levels.

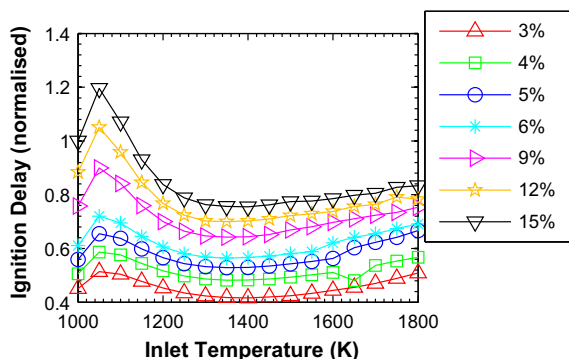


Fig. 7. Ignition delay (normalised to the no CH_2O case) for stoichiometric CH_4/H_2 with 0.5% CH_2O added across a range of inlet temperature and O_2 levels.

3.2.3. Perhydroxyl addition

The role of CH_2O on the ignition delay is clearly apparent, both from the experimental observations and the flame calculations presented. From the DNS work of Yoo et al. [18] HO_2 has also been shown to be an important precursor. Figure 8 shows the ignition delay as a function of added HO_2 to various fuel and oxidant mixtures. The addition of HO_2 reduces the ignition delay even more drastically than CH_2O . Significantly however, the relative effect is essentially the same for each mixture, in comparison to CH_2O results shown in Fig. 5. Whilst it is clear that HO_2 is a critical parameter in improving flame stabilisation, the different influence of CH_2O depending on operating conditions indicates that it is more important in understanding the differences in stabilisation behaviour for different oxidant mixtures.

3.2.4. Hydroxyl radical addition

The OH radical is a well-known marker of the flame front and can also exist in the equilibrium products of combustion. Its addition as a reactant has previously been demonstrated to reduce ignition delay times [27]. To assess the role of OH addition on ignition delay for the flame conditions of interest Fig. 9 shows a marked reduction in ignition delay for all of the mixtures. Figure 9a tends to indicate that the relative effects of OH addition are consistent for all of the CH_4/H_2 flames across the different oxidant stream conditions. The relative effect of the addition of small quantities of OH on the ignition delay is, however, dependent on the fuel and oxidant composition, as shown in Fig. 9b. Interestingly, comparing the trends of OH and CH_2O addition all four mixtures reveals the opposite trend. For instance, over the range presented in Fig. 9b the relative effects of CH_2O addition are most pronounced on CH_4 fuel and least affected by OH addition.

3.2.5. Atomic oxygen addition

The role of atomic oxygen (i.e., O) addition on the ignition delay is presented in Fig. 10. Especially for the case of cold fuel in a hot oxidiser, the O radical is expected to be an important intermediate in autoignition [19]. The general trends are very similar to OH addition. The reduction in ignition delay is similar for all the CH_4/H_2 mixtures (across the range of oxidant temperature and O_2 levels). The CH_4 case is more sensitive to atomic O addition, but the trend is similar for other oxidant mixtures (not shown). As with HO_2 and OH, the general trends of O addition to the mixture do significantly reduce the ignition delay, but these effects are largely independent of the oxidant stream properties.

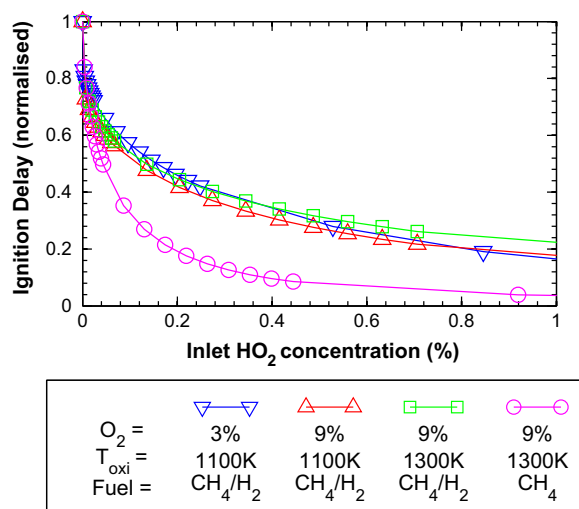


Fig. 8. Ignition delay (normalised to maximum for each condition) for various stoichiometric fuel/oxidant mixtures with the addition of HO_2 to the reactants.

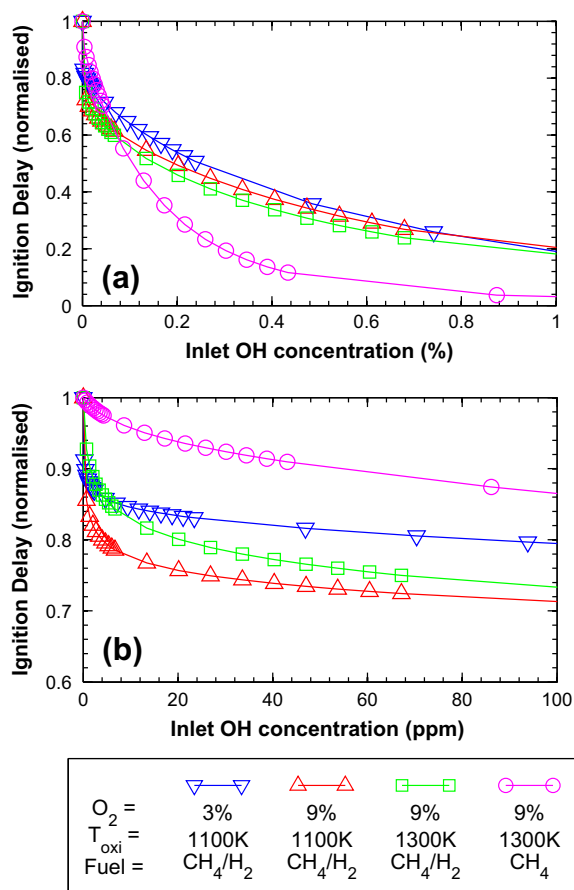


Fig. 9. Ignition delay (normalised to maximum for each condition) for various stoichiometric fuel/oxidant mixtures with the addition of OH to the reactants (shown over two different ranges of OH addition).

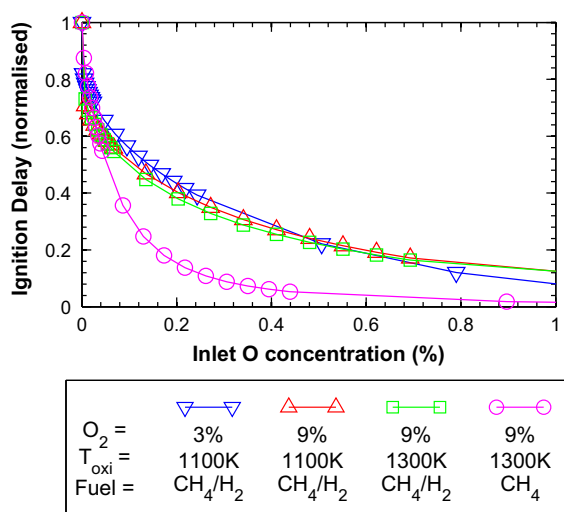


Fig. 10. Ignition delay (normalised to maximum for each condition) for various stoichiometric fuel/oxidant mixtures.

3.3. Further assessment of formaldehyde addition

3.3.1. Laminar flame speed

To further assess the role of precursor species on the flame behaviour, the effect of CH₂O addition on the laminar flame speed has been considered. For a stoichiometric CH₄/air flame the

laminar flame speed increased from 38 cm/s to 40 cm/s with the addition of 5% CH₂O to the fuel mixture (i.e., ~0.5% mole fraction of the overall mixture). This minor reduction in laminar flame speed is in contrast to the large reduction in ignition delay demonstrated previously in Fig. 5. Therefore the link between CH₂O and improved flame stability appears more closely related to the ignition delay (reactivity) rather than the laminar flame speed.

3.3.2. Opposed-flow flames with minor species addition

The evidence presented throughout this paper seems to support the proposed stabilisation theory showing a link between the increased formaldehyde (which increases with strain) and increased flame stability (through a reduction in ignition delay). With a view to understanding the role of formaldehyde on the flame structure, Fig. 11 shows one-dimensional opposed-flow laminar flame calculations for a CH₄/H₂ flame with and without the addition of 1% of formaldehyde to the fuel stream. The oxidant temperature is 1100 K and the peak strain rate is approximately 800 s⁻¹ for all cases.

Figure 11 concentrates only around the stoichiometric mixture fraction. The compositional changes to the oxidant stream give a different stoichiometric mixture fraction for each of the flames, as indicated by the arrows near the x-axis (respectively $\xi = 0.008$, 0.023 and 0.050 for the 3%, 9% and 21% O₂ cases). The solid lines (which indicate the case with formaldehyde addition to the fuel) show a sharp decline in concentration as they approach the reaction zone from the fuel-rich side as a result of consumption (noting that the initial CH₂O concentration in the fuel stream is 1%). In comparison, the dotted lines (which indicate no formaldehyde addition to the fuel) show an increase in concentration as the reaction zone is approached from the fuel-rich side: consistent with the usual trend of formaldehyde in a non-premixed flame.

Figure 11 demonstrates that the CH₂O levels at the stoichiometric mixture fraction are different for the various O₂ levels. At the stoichiometric mixture fraction for the 21% O₂ case the CH₂O level is near zero, whereas at 3% O₂ the CH₂O levels are significantly higher. This trend is consistent with the observations related to the O₂ level at the location of peak CH₂O noted previously in Fig. 1c.

Of particular note in Fig. 11 is the difference in the trend of the formaldehyde-seeded flames in comparison to the non-formaldehyde-seeded flames across the different O₂ levels. At the higher O₂ cases, with CH₂O addition the consumption rate of CH₂O on the fuel-rich side is sufficient to lower the CH₂O concentration to levels approaching the non-formaldehyde-seeded case. This is evident by the solid and dashed lines overlapping on the fuel-rich side

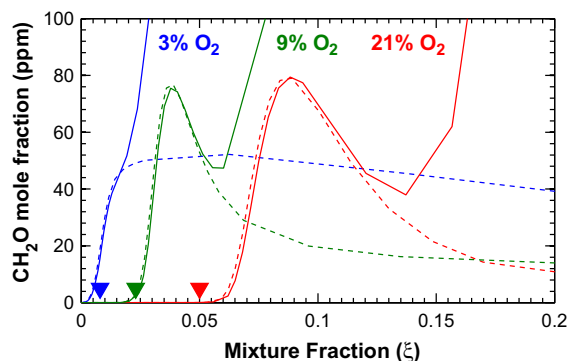


Fig. 11. Formaldehyde (CH₂O) mole fraction from opposed-flow laminar flame calculations for CH₄/H₂ fuel with 1% CH₂O added to the fuel stream (solid lines) and no formaldehyde added (dotted lines). Oxidant stream composition as per Table 1 at temperature 1100 K. Arrows along x-axis indicate stoichiometric mixture fraction for each of the cases.

of the peak CH_2O . In contrast, at 3% O_2 the two CH_2O profiles do not approach one another until the fuel-lean side of the CH_2O peak (although still fuel-rich in relation to the stoichiometric mixture fraction). These results indicate that the addition of CH_2O to the fuel stream is most likely to influence the low O_2 flames, since the CH_2O reaches further into the reaction zone.

To isolate the fate of CH_2O that is added to the fuel stream, the calculations presented in Fig. 11 for CH_4/H_2 fuel have been repeated for pure H_2 fuel. In this case the consumption of CH_2O can be analysed independent of its formation and reaction within the flame front of hydrocarbon fuels. The results are not shown graphically for brevity, but indicate the consumption of CH_2O well ahead of the reaction zone for all the O_2 cases considered. This is consistent with Fig. 11 for the 9% and 21% O_2 cases, but for H_2 fuel the trend is also the same for the 3% O_2 case (unlike for the CH_4/H_2 fuel).

To shed light on the differences between the CH_4/H_2 and H_2 fuels, the dominant consumption reactions for CH_2O for each of the fuel cases with 3% O_2 oxidant stream have been identified. (The key reactions are shown below and are labelled consistent with the GRI-Mech 3.0 kinetic mechanism.)

R58:	$\text{H} + \text{CH}_2\text{O} \rightleftharpoons \text{HCO} + \text{H}_2$
R57:	$\text{H} + \text{CH}_2\text{O} (+\text{M}) \rightleftharpoons \text{CH}_3\text{O} (+\text{M})$
R10:	$\text{O} + \text{CH}_3 \rightleftharpoons \text{H} + \text{CH}_2\text{O}$

For both CH_4/H_2 and H_2 fuel reaction R58 dominates the consumption of CH_2O added to the fuel. For the 3% O_2 case with CH_4/H_2 fuel, a low concentration of the H atom on the fuel-rich side limits the consumption of CH_2O via R58, and this enables the CH_2O added to the fuel stream to reach further into the reaction zone. This is consistent with the findings of Mardani et al. [46] who systematically investigated the role of H_2 addition to the fuel stream. It is interesting to note that CH_2O is produced via reaction R57 well upstream of the reaction zone for the case of CH_4/H_2 fuel, despite the already high concentration of CH_2O in this region. Although the consumption via R58 in this region still leads to an overall reduction in CH_2O concentration, a production of CH_2O reiterates its prevalence in these low O_2 flames. Across the reaction zone reaction R10 dominates the production, as expected. Worth noting is that for both fuels, the addition of CH_2O to the fuel stream has negligible effect on the heat release rate or its location. Furthermore, the reaction between CH_2O and OH, which is widely regarded as a marker of heat release [47–49], is less prevalent than the other reactions identified under these conditions.

For comparison, calculations have also been performed for adding CH_2O to the oxidant stream. Whilst not nominally as relevant to the lifted flame studies (where a CH_2O build-up occurs on the fuel-rich side) the results presented in Fig. 12 even more clearly show the differences between the flames. When added to the oxidant stream, for the 3% O_2 case, CH_2O exists almost over the entire mixture fraction domain, reducing only due to dilution near the fuel stream. For the 9% and 21% O_2 cases the CH_2O is seen to react well ahead of the reaction zone (even more pronounced than the case of addition to the fuel stream). These results further support the assertion that the role of CH_2O addition is heavily dependent on the oxidant stream O_2 level.

3.3.3. Experimental observations

To assess the role of CH_2O addition on the flame behaviour, experiments have also been conducted where CH_2O is added to the fuel stream. The flames are stabilised with the JHC burner [5,14]. By passing the fuel stream through a sealed vessel containing paraformaldehyde, which is heated gently, CH_2O gas is added

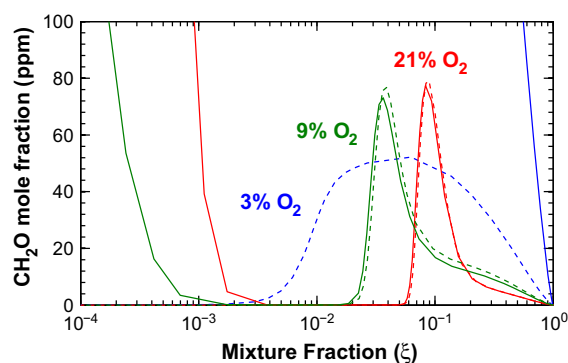


Fig. 12. Formaldehyde (CH_2O) mole fraction from opposed-flow laminar flame calculations for CH_4/H_2 fuel with 1% CH_2O added to the oxidant stream (solid lines) and no formaldehyde added (dotted lines). Oxidant stream composition as per Table 1 at temperature 1100 K.

to the jet flow. The gas lines were not heated, but were regularly cleaned to remove the products of CH_2O polymerisation [50]. The flame behaviour was observed, and recorded with photographs using a digital still camera and CH^* chemiluminescence with an intensified CCD (ICCD) camera through a 430 nm bandpass optical filter. All photographs and chemiluminescence images were each recorded with constant camera settings.

Figure 13 shows the pronounced effect of CH_2O addition on a laminar ($Re_{jet} = 1300$) natural gas flame issuing into a 12% O_2 coflow at 1300 K. Without CH_2O in the fuel stream the flame is lifted by ~ 45 mm. When CH_2O is added to the fuel the liftoff height significantly reduces, to within 5 mm from the jet exit plane. Figure 14 presents the corresponding results for a turbulent ($Re_{jet} = 7500$) flame, where CH_2O addition reduces the liftoff height from 70 mm to 40 mm. It is important to note that these observations are limited to photographs and CH^* chemiluminescence imaging. It has previously been shown that the determination of the liftoff height from visual observations can be misleading [14]. Therefore, the presented liftoff height measurements may be more appropriately referred to as the “apparent liftoff height”. Nonetheless, based on the appearance of the flame base at these flame conditions it is anticipated that the identified apparent liftoff height would correspond with the liftoff height determined from OH-LIF imaging.

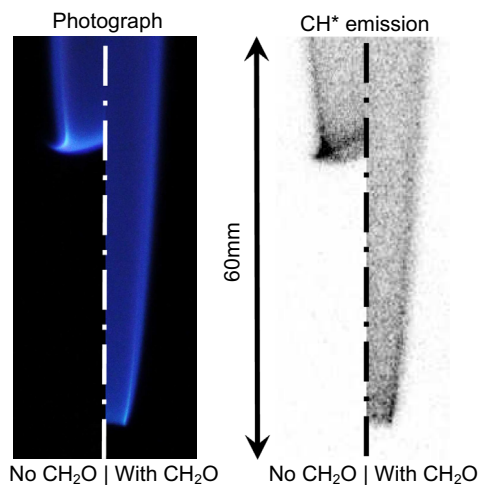


Fig. 13. Photographs and CH^* chemiluminescence images of natural gas flames (with and without CH_2O added to the fuel stream) in 12% O_2 , 1300 K coflow. $Re_{jet} = 1300$. Image height = 60 mm.

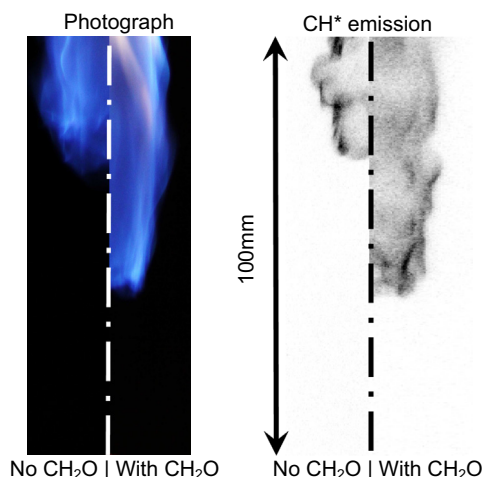


Fig. 14. Photographs and CH* chemiluminescence images of natural gas flames (with and without CH₂O added to the fuel stream) in 12% O₂, 1300 K coflow. $Re_{jet} = 7500$. Image height = 100 mm.

Worth noting is that the results presented are for a 12% O₂ coflow. Under these conditions the effects of CH₂O addition are expected to be less pronounced than under lower O₂ levels (Figs. 5 and 11). In general under MILD conditions flames stabilised on the JHC burner typically attach to the jet exit plane. This behaviour is attributed to the relatively high level of CH₂O which naturally accumulates under these conditions, such that adding CH₂O to the fuel stream has little effect in most cases. Nonetheless, experiments were also performed under 3% O₂ and 1300 K (MILD) coflow conditions, where natural gas flames typically blowoff for all jet velocities in the JHC burner. However, stable MILD combustion flames which stabilised at the jet exit plane were achieved when CH₂O added to the fuel stream (results not included in this paper due to the “flameless” nature of these MILD combustion flames). Under the 12% O₂ coflow conditions CH₂O addition had the effect of reducing the liftoff height, whereas under MILD conditions CH₂O can take a flame which was blown-off and cause it to attach to the jet exit plane. This highlights the pronounced impact that CH₂O can have on flames, especially in transitioning to MILD conditions.

4. Conclusions

Jet flames issuing into a hot and diluted oxygen coflow have previously been used to study autoignition and MILD combustion on a fundamental level. In this configuration lifted flames have been observed which demonstrate unusual behaviour, such as a reduction in liftoff height with increased velocity or decreased coflow oxygen concentration. It has previously been observed that formaldehyde is an important intermediate species in these lifted flames and it is widely accepted that formaldehyde and other precursors are indicators of autoignition. This paper has assessed the influence of this formaldehyde on the chemical kinetics, and the general trends have been demonstrated experimentally. The inclusion of small quantities of formaldehyde into the reactants has been shown to drastically affect the ignition delay. The trends of formaldehyde addition on the chemical kinetics are shown to be consistent with the experimental observations, where formaldehyde addition to the fuel stream significantly reduces the liftoff height. At low oxygen levels an increase in strain is associated with an increase in formaldehyde production. Similarly, at low oxygen levels in the coflow the addition of formaldehyde is seen to have the most influential effect on reducing the relative ignition delay.

In combination, these two processes result in jet flames issuing into a hot and highly diluted coflow being less prone to liftoff than flames with more oxygen in the coflow. These effects are even further compounded by the penetration of formaldehyde from the fuel stream reaching deeper into the reaction zone. In contrast, at the higher O₂ cases formaldehyde is less influential on the ignition delay and when added to the fuel stream in a non-premixed configuration it is consumed ahead of the reaction zone. These results therefore indicate that the unusual flame behaviour that has been observed experimentally of jet flames in a hot coflow is related to the role of formaldehyde under these conditions.

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